(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 1 195 422 A1

(12) **EUROPEAN PATENT APPLICATION**
published in accordance with Art. 158(3) EPC

(43) Date of publication:
10.04.2002 Bulletin 2002/15

(21) Application number: **00987670.7**

(22) Date of filing: **20.12.2000**

(51) Int Cl.[7]: **C09K 11/06**, H05B 33/14

(86) International application number:
PCT/JP00/09064

(87) International publication number:
WO 01/46335 (28.06.2001 Gazette 2001/26)

(84) Designated Contracting States:
AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU MC NL PT SE TR

(30) Priority: **20.12.1999 JP 36024799**
31.05.2000 JP 2000162031

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(54) THIN FILM EL DEVICE

(57) The present invention provides a thin film EL device having high electroluminescent efficiency, a low operating voltage, and a long lifetime. A thin film EL device of the present invention uses, as a luminescent layer, a charge-transport luminescent material that has, within a molecule, a portion contributing to charge transport and a portion contributing to luminescence where at least two molecular orbitals contributing to luminescent transition are localized.

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[0011] In addition, in view of the general difficulty in obtaining an organic compound having excellent electron-transport properties such as one described above, it has also been suggested that in the luminescent layer/electron-transport layer structure and in the hole-transport layer/luminescent layer/electron-transport layer structure a hole-transport luminescent material be used for the luminescent layer.

[0012] For example, Japanese Unexamined Patent Publication No. 2-250292 discloses a device having the hole-transport luminescent layer/electron-transport layer structure that uses, as the hole-transport luminescent material, [4-{2-(naphthalene-1-yl)vinyl}phenyl]bis(4-methoxyphenyl)amine or [4-(2,2-diphenylvinyl)phenyl]bis(4-methoxyphenyl) amine.

[0013] International Patent Publication No. WO96/22273 discloses a device having the hole-transport layer/hole-transport luminescent layer/electron-transport layer structure that uses, as the hole-transport luminescent material, 4,4'-bis(2,2-diphenyl-1-vinyl)-1,1'-biphenyl.

[0014] At the 1998 MRS Spring Meeting, Symposium G2.1, the hole-injecting layer/hole-transport luminescent layer/hole blocking layer/electron-transport layer structure that uses NPD as the hole-transport luminescent material was disclosed.

[0015] Further, Japanese Unexamined Patent Publications No. 10-72580 and No. 11-74079 also disclose various hole-transport luminescent materials.

[0016] Thus, using a hole-transport luminescent material as well as an electron-transport luminescent material as the luminescent material allows for the design of a wide range of materials, which in turn provides various luminous colors. However, in terms of electroluminescent efficiency, lifetimes, and so forth, it cannot be said that expectations have been met.

[0017] When devices are used in the passive-matrix line-at-a-time scanning displays, in particular, in order to attain a prescribed average luminance, peak luminance needs to be increased to very high levels. This increases the operating voltage, causing the problem of increasing power consumption as a result of power loss or the like caused by wiring resistance. Further, other problems arise, such as an increase in the cost for drive circuits and a decrease in reliability. Furthermore, devices tend to have shorter lifetimes as compared to ones used under conditions of continuous light-emission.

[0018] In addition, even with devices having high electroluminescent efficiency and relatively low operating voltages at direct current operation, when the duty ratio increases during operation, the operating voltage required to attain a prescribed average luminance is rapidly increased and also the electroluminescent efficiency itself is reduced as the operating voltage increases.

[0019] Moreover, the above-mentioned [4-{2-(naphthalene-1-yl)vinyl}phenyl] bis(4-methoxyphenyl)amine and [4-(2,2-diphenylvinyl)phenyl]bis(4-methoxyphenyl)amine, disclosed in Japanese Unexamined Patent Publication No. 2-250292, have relatively good hole-transport properties and high fluorescent yield. However, since both compounds are low-molecular-weight compounds, they suffer from the problems of low heat-resistance and particularly a short lifetime. In addition, because the compounds require luminescent dye doping, there is a problem concerning manufacturing.

[0020] The above-mentioned 4,4'-bis(2,2-diphenyl-1-vinyl)-1,1'-biphenyl, disclosed in International Patent Publication No. WO96/22273, is somewhat superior in terms of heat-resistance as compared to the above-mentioned compounds. However, since the structure of the compound is completely symmetric, the molecules easily become associated with each other, reducing electroluminescent efficiency due to microscopic crystallization and aggregation. Because of this, devices using this kind of compound are unable to obtain satisfactory lifetime when used under conditions of continuous light-emission. In addition, since the compound requires luminescent dye doping, there is a problem concerning manufacturing.

[0021] For the above-mentioned Q1-G-Q2 type compound, such as one disclosed in the 1998 MRS Spring Meeting, Symposium G2.1, besides TPD and NPD, the trimers of and the tetramers of triphenylamine have also been reported. As for their heat resistance, it has been reported that they have sufficient levels of heat resistance. However, since these compounds also have high molecular symmetry, the molecules easily become associated with each other, reducing electroluminescent efficiency due to microscopic crystallization and aggregation. Because of this, here also, devices using this kind of compound are unable to obtain satisfactory lifetimes under continuous use. Particularly when the devices are operated at high duty cycles, difficulties arise in achieving satisfactory electroluminescent efficiency and low operating voltages. In addition, since the compounds require luminescent dye doping, there is a problem concerning manufacturing.

[0022] Devices using the above-mentioned hole-transport luminescent materials disclosed in Japanese Unexamined Patent Publications No. 10-72580 and No. 11-74079 do not require luminescent dye doping, and thus are advantageous with regard to manufacturing. However, the devices have not yet achieved satisfactory electroluminescent efficiency.

operating voltage, and an extended lifetime.

[0033] The term "electron cloud of the portion contributing to charge transport" is defined herein to mean the smallest spatial extent covering 90% or more of the probability of existence of all the electrons that are related to charge transport within a molecule.

[0034] The "electron cloud of the portion contributing to luminescence" is defined herein to mean the smallest spatial extent which spatially includes at least two molecular orbitals selected from the molecular orbitals contributing to the above-mentioned luminescent transition and which covers 90% or more of the probability of existence of all the electrons that are related to luminescence within a molecule.

[0035] Specifically, the term "being localized such that the electron clouds substantially do not overlap each other" herein includes the case where there is no overlap between electron clouds that are defined by the spatial extent in which the probability of existence of all the electrons is 90% but there is overlap between electron clouds in the spatial extent in which the probability of existence of all the electrons is over 90%. As described above, the electron clouds of each portion being localized such that the electron clouds do not overlap each other are advantageous in exhibiting the function; it should be noted, however, that the case where electron clouds are localized such that the electron clouds do not overlap each other at all is not realistic, and thus such a term is used.

[0036] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the portion contributing to charge transport and the portion contributing to luminescence are connected by a carbon-carbon bond.

[0037] As with the above-mentioned structure, when the portion contributing to luminescence and the portion contributing to charge transport are connected by a carbon-carbon bond, at least two molecular orbitals contributing to luminescent transition are localized without spreading throughout the molecule, and the electron clouds of each portion are localized such that the electron clouds substantially do no overlap each other. Consequently, a device is obtained capable of exhibiting high charge transport and luminescent properties.

[0038] The term "being connected by a carbon-carbon bond" herein includes not only a direct single bond between a carbon atom contained in the portion contributing to luminescence and a carbon atom contained in the portion contributing to charge transport, but also a bond through a divalent group consisting of carbon and hydrogen atoms, such as an alkylene group and an arylene group. For such a divalent group, one having about 1 to 10 carbons is suitable. However, the "carbon-carbon bond" does not include a bond through nitrogen atoms or the like, a direct carbon-carbon double bond, and a direct carbon-carbon triple bond because these may hinder the localization of molecular orbitals.

[0039] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the charge-transport luminescent material is a compound having an asymmetric and nonplanar molecular structure.

[0040] As with the above-mentioned structure, when the molecular structure is asymmetric and nonplanar, amorphous characteristics and non-associating properties are exhibited, and therefore quenching due to the interaction between each of the portions contributing to luminescence of adjacent molecules or the like can be suppressed. As a result, a device is obtained that has high electroluminescent efficiency.

[0041] The term "asymmetric and nonplanar" is defined herein to mean that the molecular structure at its most stable state is not symmetric with respect to a point, a line, or a plane, but is three dimensional.

[0042] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the portion contributing to luminescence is present within the luminescent layer at 1×10^{20} to 1×10^{21} per 1 cm^3.

[0043] As with the above-mentioned structure, when the portion contributing to luminescence is present within the luminescent layer at a specific density, a device is obtained that achieves high luminance with high electroluminescent efficiency. This can be explained by the fact that when the density of the portion contributing to luminescence is too low, sufficient luminance tends not to be obtained; on the contrary, when the density is too high, quenching occurs due to the interaction between the portions contributing to luminescence, and thus electroluminescent efficiency tends to be degraded.

[0044] Here, the number of the portions contributing to luminescence is counted per portion; for example, when the charge-transport luminescent material has two portions contributing to luminescence within a molecule, the number of the portions contributing to luminescence per unit area equals a value that is double the number of molecules per unit area.

[0045] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the volume ratio of the portion contributing to luminescence is lower than that of the portion contributing to charge transport.

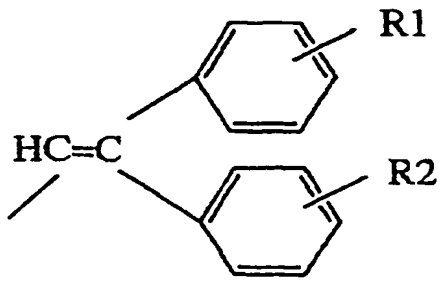
[0046] As with the above-mentioned structure, when the volume ratio of the portion contributing to luminescence is lower than that of the portion contributing to charge transport, the possibility of quenching due to the interaction between the portions contributing to luminescence is suppressed. Consequently, a device is obtained that achieves high electroluminescent efficiency.

[0047] The term "volume ratio" is herein defined as the ratio of the volume occupied by the portion contributing to luminescence and the like to the total volume of a molecule having the portion contributing to luminescence and the like.

a hole-electron recombination is increased. Thus, a thin film EL device is obtained that achieves high electroluminescent efficiency.

[0059] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the X in the general formula (1) is a substituent represented by the following general formula (2):

(2)

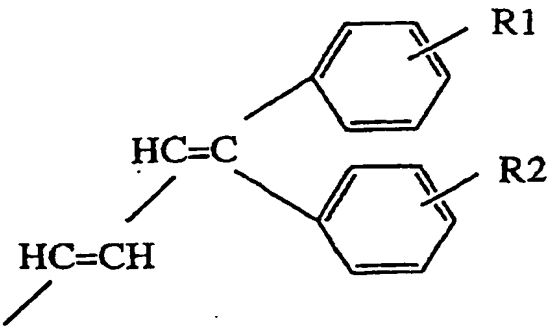


where R1 and R2 may be the same or different, and each independently represents a hydrogen atom or an alkyl group.

[0060] As with the above-mentioned structure, when the X in the general formula (1) is a bulky substituent such as one represented by the general formula (2), this portion becomes twisted and thus the molecules of the hole-transport luminescent material become asymmetric and nonplanar. Thus, molecular association, crystallization, and the like are less likely to occur, resulting in a device achieving high electroluminescent efficiency

[0061] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the X in the general formula (1) is a substituent represented by the following general formula (3):

(3)



where R1 and R2 may be the same or different, and each independently represents a hydrogen atom or an alkyl group.

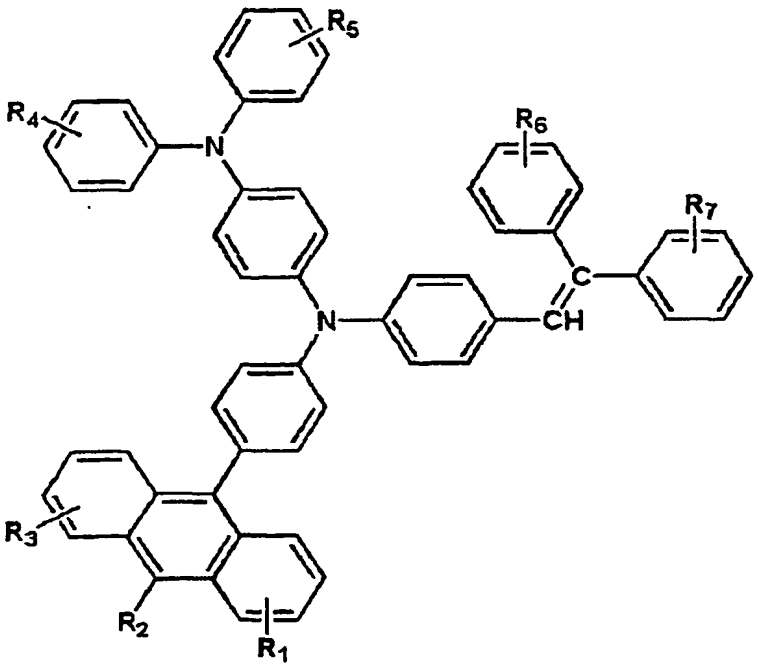
[0062] The substituent represented by the above-mentioned general formula (3) is a bulky substituent in which a vinyl group is bonded to a substituent represented by the above-mentioned formula (2). Thus, molecular association, crystallization, and the like are less likely to occur, resulting in a device achieving high electroluminescent efficiency and so forth.

[0063] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the X in the general formula (1) is a substituent represented by the following general formula (4):

and a low operating voltage.

[0073] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the hole-transport luminescent material is a compound represented by the following general formula (6):

(6)



where R4, R5, R6, and R7 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group; and R1, R2, and R3 may be the same or different, and each independently represents a hydrogen atom or an electron-donating substituent.

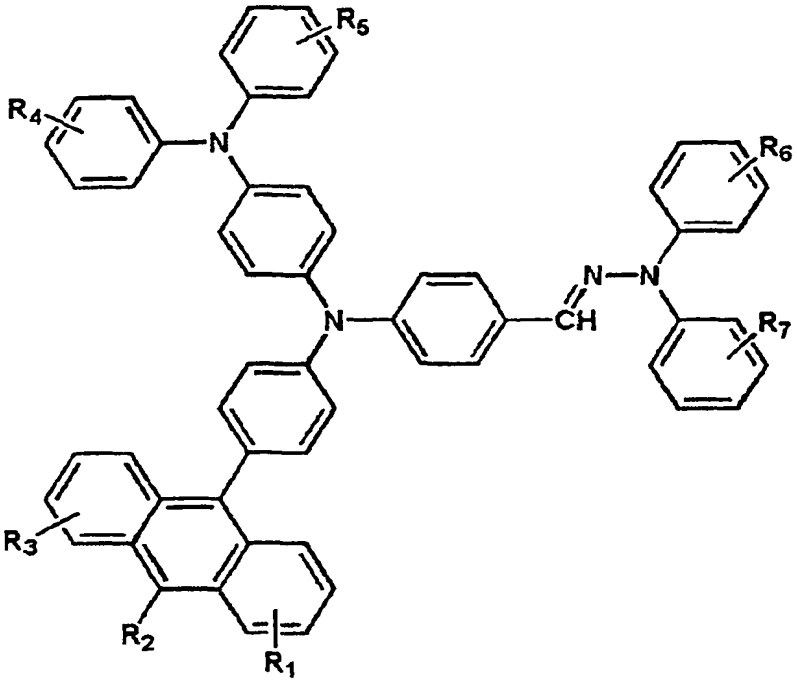
[0074] In the above-mentioned compound, the portion contributing to hole transport is of a tetraphenyl-p-phenylenediamine skeleton and the portion contributing to luminescence is an anthryl group. One phenyl group of diphenylamine is substituted with the above-mentioned anthryl group and the other is substituted with a substituted or unsubstituted 2,2-diphenylvinyl group. Such compound has a portion contributing to luminescence where at least two molecular orbitals contributing to luminescent transition are localized, and an electron cloud of the portion contributing to luminescence and a molecular cloud of the portion contributing to hole transport are localized such that the electron cloud and the molecular cloud do not overlap each other. Further, since a bulky substituent, a 2,2-diphenylvinyl group, is bonded, this portion becomes twisted and thus the molecules become asymmetric and nonplanar. Thus, a thin film EL device is obtained that achieves high electroluminescent efficiency, a low operating voltage, and an extended lifetime even when the device is operated at a wide range of operating conditions, from a direct current to high duty cycles.

[0075] A compound represented by the above-mentioned general formula (6) may be (4-{[4-(2,2-diphenylvinyl)phenyl][4-(9-anthryl)phenyl]amino}phenyl) diphenylamine, (4-{[4-(2,2-diphenylvinyl)phenyl][4-(10-methoxy(9-anthryl))phenyl]amino}phenyl)diphenylamine, or the like.

[0076] In this patent specification, the names of compounds used herein were named so as to conform to IUPAC nomenclature rules. Specifically, the compounds were named using Chemistry 4-D Draw (available from ChemInnovation Software, Inc.) based on the structural formulae for each compound.

[0077] According to another aspect of the present invention the above-mentioned thin film EL device may be such that a compound represented by the following general formula (7) is used as the hole-transport luminescent material:

(8)



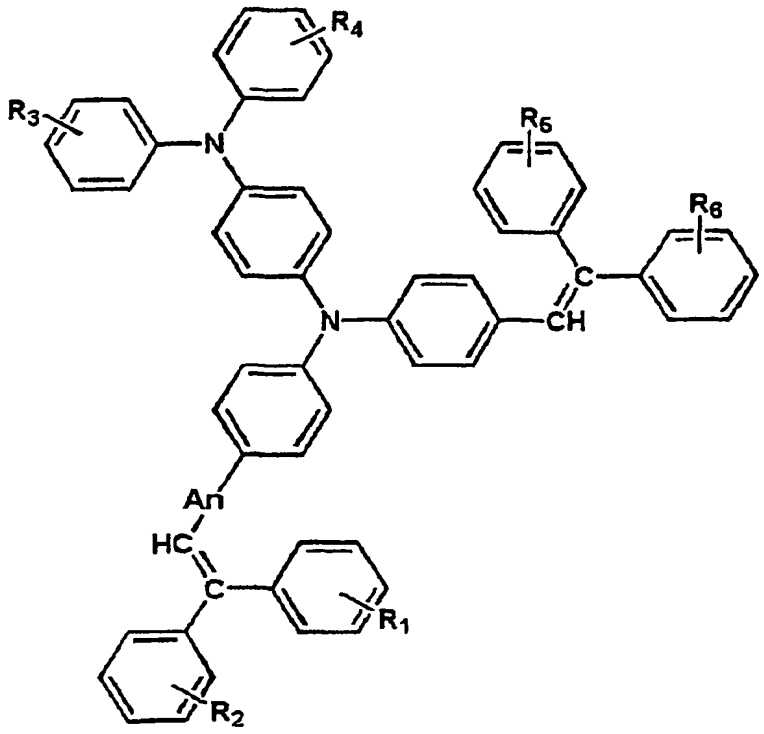
where R4, R5, R6, and R7 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group; and R1, R2, and R3 may be the same or different, and each independently represents a hydrogen atom or an electron-donating substituent.

[0081] The above-mentioned hole-transport luminescent material includes an anthryl group, corresponding to the portion contributing to luminescence, and a tetraphenyl-p-phenylenediamine skeleton, corresponding to the portion contributing to hole transport, and further includes a bulky substituent, a substituted or unsubstituted 2-aza-2-diphenylaminovinyl group. Thus, a thin film EL device is obtained that achieves particularly high electroluminescent efficiency, a low operating voltage, and an extended lifetime even when the device is operated at various operating conditions.

[0082] A compound represented by the above-mentioned general formula (8) may be [4-({4-[2-aza-2-(diphenylamino)vinyl]phenyl}{4-(9-anthryl)phenyl} amino)phenyl]diphenylamine, [4-({4-[2-aza-2-(diphenylamino)vinyl]phenyl} {4-(10-methoxy(9-anthryl))phenyl}amino)phenyl]diphenylamine, or the like.

[0083] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the hole-transport luminescent material is a compound represented by the following general formula (9):

(10)



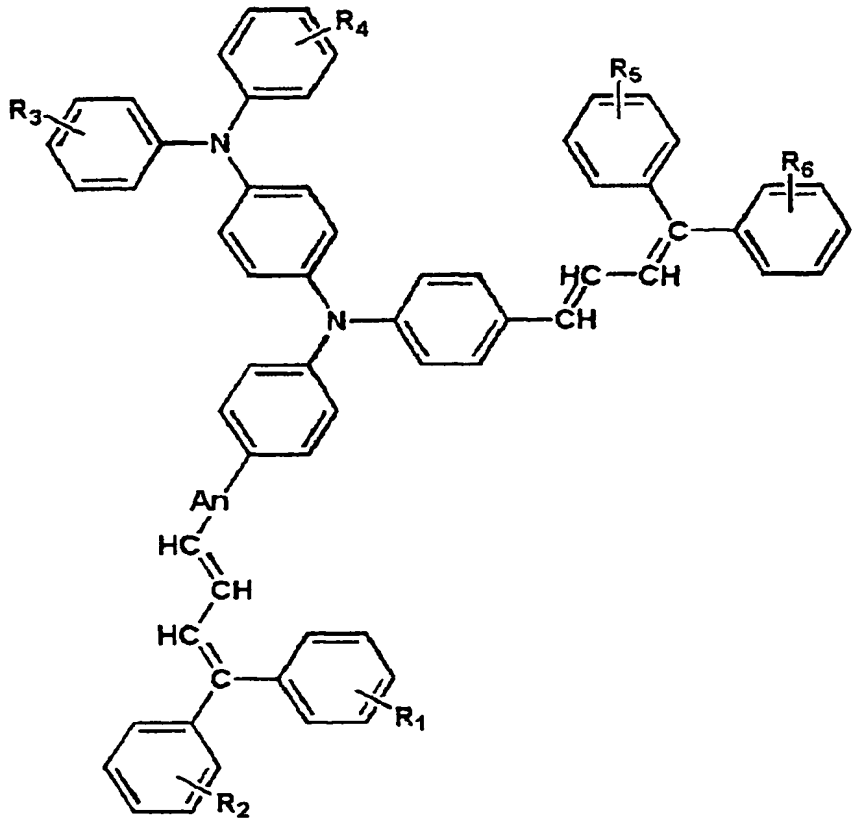
where R1, R2, R3, R4, R5, and R6 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group; and An represents an arylene group composed of two or more substituted or unsubstituted fused rings.

[0087] The above-mentioned hole-transport luminescent material includes an arylene group composed of two or more fused rings, corresponding to the portion contributing to luminescence, and a tetraphenyl-p-phenylenediamine skeleton, corresponding to the portion contributing to hole transport. In addition, the material includes two bulky substituents, substituted or unsubstituted 2,2-diphenylvinyl groups. Thus, a thin film EL device is obtained that achieves particularly high electroluminescent efficiency, a low operating voltage, and an extended lifetime even when the device is operated at various operating conditions.

[0088] A compound represented by the above-mentioned general formula (10) may be [4-({4-[10-(2,2-diphenylvinyl)(9-anthryl)]phenyl}[4-(2,2-diphenylvinyl)phenyl]amino)phenyl]diphenylamine, [4-({4-[10-(2,2-diphenylvinyl)(9-anthryl)]phenyl}{4-(2,2-diphenylvinyl)phenyl}amino) phenyl]bis(4-methoxyphenyl)amine, or the like.

[0089] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the hole-transport luminescent material is a compound represented by the following general formula (11):

(12)



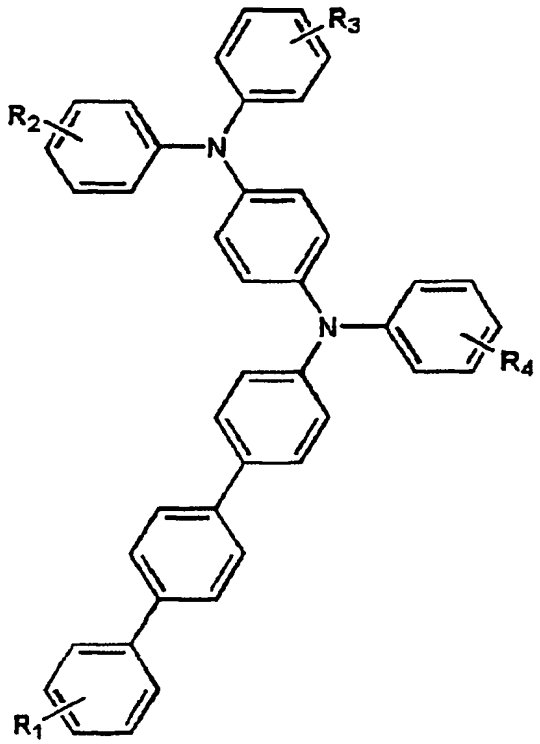
where R1 and R2 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group; and An represents an arylene group composed of two or more substituted or unsubstituted fused rings.

[0093] The above-mentioned hole-transport luminescent material includes an arylene group composed of two or more fused rings, corresponding to the portion contributing to luminescence, and a tetraphenyl-p-phenylenediamine skeleton, corresponding to the portion contributing to hole transport. In addition, the hole-transport luminescent material is substituted with two bulky substituents, substituted or unsubstituted 4,4-diphenylbuta-1,3-dienyl groups. Thus, a thin film EL device is obtained that achieves particularly high electroluminescent efficiency, a low operating voltage, and an extended lifetime even when the device is operated at various operating conditions.

[0094] A compound represented by the above-mentioned general formula (12) may be [4-({4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)]phenyl}[4-(4,4-diphenylbuta-1,3-dienyl)phenyl] amino)phenyl] diphenylamine, [4-({4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)]phenyl}{4-(4,4-diphenylbuta-1,3-dienyl)phenyl}amino)phenyl]bis(4-methoxy-phenyl)amine, or the like.

[0095] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the hole-transport luminescent material is a compound represented by the following general formula (13):

(14)



where R4 represents a hydrogen atom, an alkyl group, an alkoxy group, or an aralkyl group; and R1, R2, and R3 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group.

[0099] The above-mentioned hole-transport luminescent material includes a terphenyl group, corresponding to the portion contributing to luminescence, and a tetraphenyl-p-phenylenediamine skeleton, corresponding to the portion contributing to hole transport. This material also includes a terphenyl group, which is the portion contributing to luminescence, and thus a thin film EL device is obtained that achieves high electroluminescent efficiency, a low operating voltage, and an extended lifetime even when the device is operated at various operating conditions.

[0100] A compound represented by the above-mentioned general formula (14) may be [4-(diphenylamino)phenyl] [4-(4-phenylphenyl)phenyl]phenylamine, [4-{bis(4-methoxyphenyl)amino}phenyl][4-{4-(4-methoxyphenyl)phenyl} phenyl][4-(1-methyl-1-phenylethyl)phenyl]amine, or the like.

[0101] According to another aspect of the present invention the above-mentioned thin film EL device may be such that the hole-transport luminescent material is a compound represented by the following general formula (15):

one made of polyester. A preferable thickness of the substrate is 0.5 to 1.0 mm thickness in terms of strength and weight.

[0108] For the hole-injecting electrode 2, there are no particular limitations as long as it functions as the anode and is capable of injecting holes into the hole-transport layer 3. It should be noted, however, that either the hole-injecting electrode 2 or the electron-injecting electrode 6 described below is made to have transparency to extract the emitted light to the outside and it is often the case that normally the hole-injecting electrode 2 is made to be a transparent electrode. In this case, an ITO (indium tin oxide) film is usually used. In forming an ITO film, in order to ensure a high degree of transparency and a low resistivity, such film-forming techniques as sputtering, electron beam evaporation, or ion plating are employed. The formed ITO film may be given various post-treatments to control its resistivity and shape. The film thickness is determined mainly from sheet resistance and visible light transmittance; however, since thin film EL devices have relatively high operating current densities, in order to reduce the sheet resistance, films are usually formed to be a thickness of 100 nm or more, generally 100 to 150 nm. In addition to an ITO film, which is a transparent electrode, it is also possible to use various improved transparent conductive layers, such as an In_2O_3-ZnO transparent conductive electrode (IDIXO available from Idemitsu Kosan Co.,Ltd.), or a coating film of a transparent conductive coating in which conductive powder particles are dispersed.

[0109] For the electron-injecting electrode 6, an electrode that is composed of an alloy of a low work function metal with a low electron injection barrier and a relatively high work function, stable metal is used; for example, an MgAg alloy proposed by Tang et al. as described in the Background Art or an AlLi alloy. In addition, it is possible to use various structures of electrodes, such as a multi-layer cathode composed of a Li thin film and an Al film, which is thicker than the Li thin film, or a multi-layer cathode composed of a LiF film and an Al film.

[0110] The hole-transport layer 3 and the electron-transport layer 5 sandwiched between the above-mentioned hole-injecting electrode 2 and the electron-injecting electrode 6 have no particular limitations, and thus are formed using any type of material known in the prior art. For the hole-transport layer 3, a layer is used composed of a material having hole-transport properties, such as TPD or NPD described above. It is also possible to use a specific material-blended type hole-transport layer, which is disclosed in Japanese Unexamined Patent Publication No. 11-260559. For the electron-transport layer 5, a layer composed of various materials having electron-transport properties is used; for example, an aluminum quinoline complex, such as the above-mentioned tris(8-quinolinolato)aluminum (Alq3), or various compounds, such as all kinds of oxadiazole derivatives or phenanthroline derivatives, can be widely used.

[0111] Next, for the luminescent layer 4, which is the most significant feature of the present invention, the charge-transport luminescent material is used having a portion contributing to charge transport and a portion contributing to luminescence where at least two molecular orbitals (for example, HOMO and LUMO) contributing to luminescent transition are localized. The portion contributing to charge transport may be of, for example, a tetraphenyl phenylenediamine skeleton or the like. With this skeleton, generally, higher electroluminescent efficiency and longer lifetimes than triphenylamine dimer (TPD and the like), so-called Q1-G-Q2 structure, are achieved. The portion contributing to luminescence may be of, for example, an anthracene skeleton or the like. With this skeleton, particularly good electroluminescent efficiency and high charge transport properties are achieved, and in addition low operating voltages and low power consumption are achieved.

[0112] Among the above-mentioned charge-transport luminescent materials, a material is particularly suitable in which an electron cloud of the portion contributing to charge transport and an electron cloud of the portion contributing to luminescence are localized such that the electron clouds substantially do not overlap each other. When this kind of material is used, charge transport properties and luminescent properties can be exhibited separately, and thus an excellent thin film EL device is obtained. In addition, when a carbon atom of the portion contributing to charge transport and a carbon atom of the portion contributing to luminescence are connected by a carbon-carbon bond, the electron clouds are localized individually such that the electron clouds substantially do not overlap each other, ensuring an excellent thin film EL device.

[0113] The luminescent layer 4 is formed using such charge-transport luminescent materials by various film-forming techniques such as vapor deposition. Since the luminescent layer 4 of the present invention achieves high electroluminescent efficiency, normally, it is not necessary to dope the layer with luminescent dyes. Thus, a thin film EL device suitable for mass production is obtained.

[0114] A hole-transport luminescent material may be a compound that is represented by the above-mentioned general formula (1). Above all, a compound represented by the above-mentioned general formulae (6) to (15) is preferable, and a compound represented by the general formulae (6) to (13) is more preferable.

[0115] For substituted or unsubstituted aryl groups represented by Ar1 and Ar2 in the above-mentioned formula (1), preferable examples thereof include the following: for an unsubstituted aryl group, one having 6 to 20 carbons is suitably used. Specifically, a phenyl group, a biphenyl group, a terphenyl group, a naphthyl group, an anthryl group, a phenanthryl group, a fluorenyl group, and the like, are suitable. A substituted aryl group may be one in which the above-mentioned unsubstituted aryl group is substituted with, for example, an alkyl group having 1 to 10 carbons, an alkoxy group having 1 to 10 carbons, or the like.

[0116] For a substituted or unsubstituted arylene group represented by Ar3 in Formula (1), preferable examples

of 0.2 Torr (1 Torr = 133.322 Pa), and a high frequency output of 300 W.

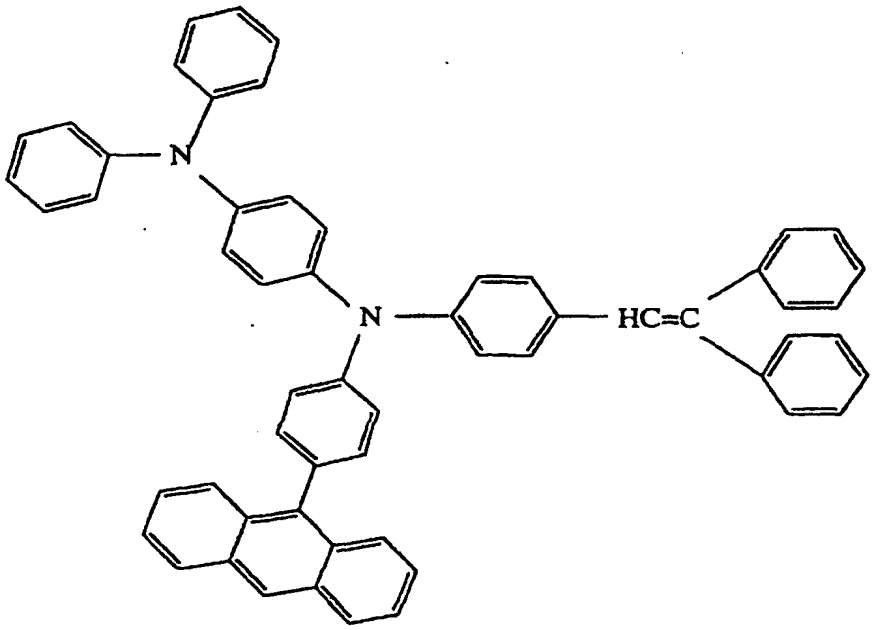
[0128] The hole-injecting electrode-coated substrate thus prepared was placed in the vacuum chamber of a vacuum evaporator. The vacuum evaporator used here is one in which a main pumping system of a commercially available vacuum evaporator (Model EBV-6DA, available from ULVAC Japan, Ltd.) is modified. In this system, the main pumping system is a turbo molecular pump with a pumping speed of 1500 liters/min (TC1500, available from Osaka Vacuum, Ltd.) and has an ultimate vacuum of about 1×10^{-6} Torr or less, and all vapor depositions were carried out in the range of 2 to 3×10^{-6} Torr. In addition, all vapor depositions were carried out by connecting a tungsten boat for resistance-heated evaporation to the DC power supply (PAK10-70A, available from Kikusui Electronics Corporation).

[0129] The hole-injecting electrode-coated substrate was placed in the vacuum chamber of a system such as one described above. Onto the substrate, N,N'-bis (4'-diphenylamino-4-biphenylyl)-N,N'-diphenylbenzidine (TPT, available from Hodogaya Chemical Co., Ltd.) and 4-N,N-diphenylamino-α-phenylstilbene (PS) were co-deposited at deposition rates of 0.3 (nm/s) and 0.01 (nm/s), respectively, to form a material-blended type hole-transport layer with a thickness of about 80 (nm).

[0130] Then, (4-{[4-(2,2-diphenylvinyl)phenyl](4-(9-anthryl)phenyl)amino} phenyl)diphenylamine (hereafter referred to as "PPDA-PS-A"), which is a hole-transport luminescent material, was vapor deposited at a deposition rate of 0.3 nm/s to form a hole-transport luminescent layer with a thickness of about 40 nm.

[0131] Here, the PPDA-PS-A was a compound represented by the following chemical formula (16) and was obtained by synthesizing as follows.

(16)



[0132] As a starting material, N-acetyl-1,4-phenylenediamine (TCI Catalog No. A0106, 2250 yen/25 g) was prepared and underwent the Ullmann reaction with iodobenzene. The resulting substance was then hydrolyzed, and further underwent the Ullmann reaction with 9-(4-iodophenyl) anthracene.

[0133] Thereafter, the resultant was formylated by the Vilsmeier reaction as shown in the following reaction formula (17). Here, for a solvent used for the reaction, it is possible to use DMF to obtain high reactivity, but in order to enhance reaction selectivity and increase the proportion of the target compound, N-methylformanilide was used. In addition since the Vilsmeier reaction is electrophilic addition, a carbon having the highest HOMO electron density became a reactive site, and thus the para position of the benzene ring, which is directly bonded to the nitrogen, was formylated. After the formylation, the resultant was thoroughly isolated by column chromatography and thus the target compound was extracted.

ditions with ambient temperature and humidity. It should be noted that the operating voltage was such a level obtained at a luminance level of 1000 (cd/m^2).

[0141] As for the lifetime, under the same conditions as described above, a continuous light-emission test was conducted by operating the device at a DC constant-current level that provides an initial luminance of 1000 (cd/m^2). Then, the lifetime, which is defined as the time taken for luminance to decrease by half (500 cd/m^2), was evaluated.

[0142] Here, the device was operated with DC constant-current by using a DC constant-current power supply (Multi-Channel Current Voltage Controller TR6163, available from Advantest Corporation). The luminance was measured using a luminance meter (Topcon luminescence meter BM-8, available from Topcon Corporation).

(Evaluation of the device when operated with pulsed constant-current)

[0143] The electroluminescent efficiency (cd/A) and the operating voltage (V) were evaluated by, under the same conditions as described above, operating the device with pulsed constant-current. It should be noted that the operating voltage was such a level obtained at an average luminance of 270 (cd/m^2).

[0144] As for the lifetime, under the same conditions as described above, a continuous light-emission test was conducted by operating the device at a pulsed constant-current level that provides an average luminance of 270 (cd/m^2). Then, the lifetime, which is defined as the time taken for luminance to decrease by half (135 cd/m^2), was evaluated.

[0145] Here, the device was operated with pulsed constant-current by using a pulsed constant-current drive circuit. Operating conditions were such that the pulse frequency was 100 Hz (10 ms), the duty ratio was 1/240 (a pulse width of 42 μs), and the pulse waveform was a square wave. Under these operating conditions, evaluations were made by operating the device at various pulsed current levels. The luminance was measured using a luminance meter (Topcon luminescence meter BM-8, available from Topcon Corporation).

[0146] In addition to the above evaluations, the quality of luminescent images, such as uneven luminance and dark spots (non-light-emitting portions), was observed while the device was being operated to emit light by using an optical microscope with 50 times magnification.

[0147] The results of these evaluations are provided in Table 1 to be presented later.

(Example 2)

[0148] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that (4-{[4-(2,2-diphenylvinyl)phenyl] [4-(10-methoxy(9-anthryl))phenyl]amino}phenyl)diphenylamine (PPDA-PS-AM) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 3)

[0149] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that (4-{[4-(4,4-diphenylbuta-1,3-dienyl)phenyl](4-(9-anthryl)phenyl)amino}phenyl)diphenylamine (PPDA-PB-A) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 4)

[0150] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that (4-{[4-(4,4-diphenylbuta-1,3-dienyl)phenyl][4-(10-methoxy)(9-anthryl)]pheny}amino)phenyl) diphenylamine (PPDA-PB-AM) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 5)

[0151] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-({4-[2-aza-2-(diphenylamino) vinyl]phenyl}(4-(9-anthryl)phenyl)amino)phenyl]diphenylamine (PPDA-PH-A) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 13)

[0159] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-({4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)]phenyl}[4-(4,4-diphenylbuta-1,3-dienyl)phenyl]amino) phenyl] diphenylamine (PPDA-PB-APB) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 14)

[0160] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-({4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)]phenyl}[4-(4,4-diphenylbuta-1,3-dienyl)phenyl]amino) phenyl] bis(4-methoxyphenyl)amine (M2PPDA-PB-APB) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 15)

[0161] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that {4-[bis(4-(9-anthryl)phenyl) amino]phenyl}diphenylamine (PPDA-A2) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 16)

[0162] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-(bis{4-[10-(2,2-diphenylvinyl) (9-anthryl)]phenyl}amino)phenyl]diphenylamine (PPDA-APS2) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 17)

[0163] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-(bis{4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)]phenyl}amino)phenyl]diphenylamine (PPDA-APB2) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 18)

[0164] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-(bis{4-[10-(fluorene-9-ylidenmethyl)(9-anthryl)]phenyl}amino)phenyl]diphenylamine (PPDA-AFM2) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 19)

[0165] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-(diphenylamino)phenyl][4-(4-phenylphenyl)phenyl]phenylamine (TPPDA) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Example 20)

[0166] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-{bis(4-methoxyphenyl)amino} phenyl][4-{4-(4-methoxyphenyl)phenyl}phenyl][4-(1-methyl-1-phenylethyl) phenyl]amine (MTPPDA) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A.

(Comparative Example 6)

[0174] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-(diphenylamino)phenyl] diphenylamine (TPPDA) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Comparative Example 7)

[0175] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-{(4-phenylphenyl) phenylamino}phenyl](4-phenylphenyl)phenylamine (DPBBPDA) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

(Comparative Example 8)

[0176] A sample of the thin film EL device was fabricated in a similar manner to that described in Example 1, except that [4-{bis(4-phenylphenyl)amino} phenyl]bis(4-phenylphenyl)amine (TBPDA) was used for the formation of the hole-transport luminescent layer in place of the PPDA-PS-A. The sample was evaluated in a similar manner to that described in Example 1. The results of the evaluations are provided in Table 1 to be presented later.

efficiency and achieve luminescence with good visibility with low operating voltages and self-emission. In addition, the continuous light-emission tests revealed that the devices showed little degradation in luminance, had no defects such as dark spots or uneven luminance, and were capable of operating stably over an extremely long period of time.

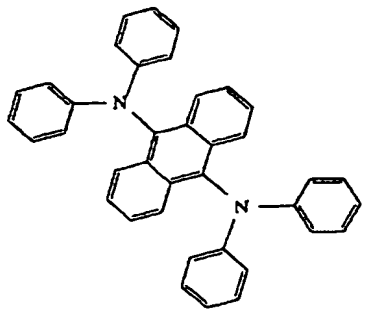
[0178] Particularly, even in pulsed-operation corresponding to the actual panel operation, the devices have high electroluminescent efficiency and low operating voltages. In addition, the continuous light-emission tests revealed that the devices showed little degradation in luminance, had no defects such as dark spots or uneven luminance, and were capable of operating stably over an extremely long period of time.

[0179] Further, the devices of Examples 1 to 18 achieve higher electroluminescent efficiency, lower operating voltages, and longer lifetimes as compared to those of Examples 19 to 22. This may be explained by the fact that in the devices of Example 1 to 18 the electron clouds of the portions contributing to hole transport and the electron clouds of the portions contributing to luminescence are localized such that the electron clouds substantially do not overlap each other.

[0180] In Table 1 above, the constituent compounds of the devices of each example and comparative example are represented in an abbreviated form as follows:

TPT indicates N,N'-bis(4'-diphenylamino-4-biphenylyl)-N,N'-diphenylbenzidine;
PS indicates 4-N,N-diphenylamino-αa-phenylstilbene;
The PPDA-PS-A indicates (4-{[4-(2,2-diphenylvinyl)phenyl](4-(9-anthryl) phenyl)amino}phenyl)diphenylamine;
PPDA-PS-AM indicates (4-{[4-(2,2-diphenylvinyl)phenyl][4-(10-methoxy(9-anthryl))phenyl] amino}phenyl)diphenylamine;
PPDA-PB-A indicates (4-{[4-(4,4-diphenylbuta-1,3-dienyl)phenyl](4-(9-anthryl)phenyl)amino}phenyl)diphenylamine;
PPDA-PB-AM indicates (4-{[4-(4,4-diphenylbuta-1,3-dienyl)phenyl][4-(10-methoxy)(9-anthryl)]phenyl}amino)phenyl)diphenylamine;
PPDA-PH-A indicates [4-({4-[2-aza-2-(diphenylamino)vinyl]phenyl}(4-(9-anthryl)phenyl)amino)phenyl] diphenylamine;
PPDA-PH-AM indicates [4-({4-[2-aza-2-(diphenylamino)vinyl]phenyl}(4-(10-methoxy(9-anthryl))phenyl)amino) phenyl]diphenylamine;
PPDA-FM-A indicates (4-{[4-(fluorene-9-ylidenemethyl)phenyl][4-(9-anthryl) phenyl]amino}phenyl)diphenylamine;
PPDA-FM-AM indicates (4-{[4-(fluorene-9-ylidenemethyl)phenyl][4-(10-methoxy(9-anthryl))phenyl]amino}phenyl) diphenylamine;
PPDA-PS-APS indicates [4-({4-[10-(2,2-diphenylvinyl)(9-anthryl)]phenyl}[4-(2,2-diphenylvinyl)phenyl] amino)phenyl]diphenylamine;
M2PPDA-PS-APS indicates [4-({4-[10-(2,2-diphenylvinyl)(9-anthryl)]phenyl} [4-(2,2-diphenylvinyl)phenyl]amino) phenyl]bis(4-methoxyphenyl)amine;
PPDA-FM-AFM indicates [4-({4-[10-(fluorene-9-ylidenemethyl)(9-anthryl)] phenyl}[4-(fluorene-9-ylidenemethyl) phenyl]amino)phenyl]diphenylamine;
M2PPDA-FM-AFM indicates [4-({4-[10-(fluorene-9-ylidenemethyl)(9-anthryl)]phenyl}[4-(fluorene-9-ylidenemethyl)phenyl]amino)phenyl]bis(4-methoxyphenyl)amine;
PPDA-PB-APB indicates [4-({4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)] phenyl} [4-(4,4-diphenylbuta-1,3-dienyl)phenyl] amino)phenyl] diphenylamine;
M2PPDA-PB-APB indicates [4-({4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)]phenyl}[4-(4,4-diphenylbuta-1,3-dienyl)phenyl]amino)phenyl]bis(4-methoxyphenyl)amine;
PPDA-A2 indicates {4-[bis(4-(9-anthryl)phenyl)amino]phenyl} diphenylamine;
PPDA-APS2 indicates [4-(bis{4-[10-(2,2-diphenylvinyl)(9-anthryl)]phenyl} amino)phenyl]diphenylamine;
PPDA-APB2 indicates [4-(bis{4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)] phenyl}amino)phenyl]diphenylamine;
PPDA-AFM2 indicates [4-(bis{4-[10-(fluorene-9-ylidenemethyl)(9-anthryl)] phenyl}amino)phenyl]diphenylamine,
TPPDA indicates [4-(diphenylamino)phenyl][4-(4-phenylphenyl)phenyl) phenylamine;
MTPPDA indicates [4-{bis(4-mthoxyphenyl)amino}phenyl][4-{4-(4-methoxyphenyl)phenyl}phenyl][4-(1-methyl-1-phenylethyl)phenyl]amine;
T2PPDA indicates [4-(diphenylamino)phenyl][bis{4-(4-phenylphenyl) phenyl}]amine;
MT2PPDA indicates [4-{bis(4-methoxyphenyl)amino}phenyl]bis[4-{4-(4-methoxyphenyl)phenyl}phenyl]amine;
DANS indicates [4-{2-(naphthalene-1-yl)vinyl}phenyl]bis(4-methoxyphenyl) amine;
MDAPS indicates [4-(2,2-diphenylvinyl)phenyl]bis(4-methoxyphenyl)amine;
DPVBi indicates 4,4'-bis(2,2-diphenyl-1-vinyl)-1,1'-biphenyl;
TPD indicates N,N'-bis(3-methylphenyl)-N,N'-diphenylbenzidine;

(20)



(Table 2)

	Absorption Wavelength (nm)	Oscillator Strength
Chemical Formula (18)	343.1	0.466
Chemical Formula (19)	383.3	0.836
Chemical Formula (20)	376.8	0.390

[0183] Table 2 shows that the compounds represented by the above-mentioned chemical formulae (18) and (19) have higher oscillator strengths than the compound represented by the chemical formula (20). The oscillator strength and the electroluminescent efficiency are correlated, that is to say, when the oscillator strength is high, the electroluminescent efficiency is high. Thus, a device using the compound represented by the chemical formula (18) or (19) as the luminescent material achieves high electroluminescent efficiency.

[0184] In addition, the compound represented by the chemical formula (19) has higher oscillator strength than the compound represented by the chemical formula (18). The compound represented by the chemical formula (19) is one in which a methoxy group (an electron-donating substituent) is directly bonded to an anthracene skeleton (a portion contributing to luminescence) of the compound represented by the chemical formula (18). Consequently, a device using a compound, in which an electron-donating substituent is directly bonded to a portion contributing to luminescence, achieves higher electroluminescent efficiency.

INDUSTRIAL APPLICABILITY

[0185] As has been described thus far, according to the present invention, a thin film EL device uses, as the charge-transport luminescent material, a compound represented by the above mentioned general formula (1) that has a portion contributing to charge transport and a portion contributing to luminescence where at least two molecular orbitals contributing to luminescent transition are localized. Thus, it is possible to provide self-luminous devices with excellent visibility that exhibit high electroluminescent efficiency, low operating voltages, and longer lifetimes even when operated at various operating voltages. In addition, the continuous light-emission tests revealed that the devices showed little degradation in luminance and were capable of operating stably with low power consumption over an extremely long period of time.

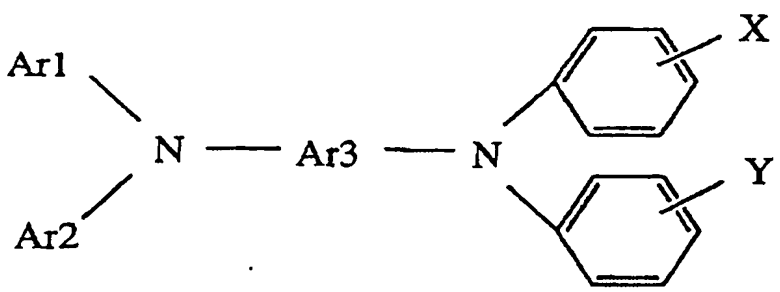
[0186] Furthermore, even in pulsed operation corresponding to the actual operation of the passive matrix panel, the devices have low operating voltages, high efficiency, and high reliability and are capable of operating stably with low power consumption over an extremely long period of time.

[0187] Thus, the present invention is useful in fields such as various kinds of light sources used for self-luminous flat panel displays, telecommunications, lighting, and other applications.

Claims

1. A thin film EL device comprising at least:

(1)

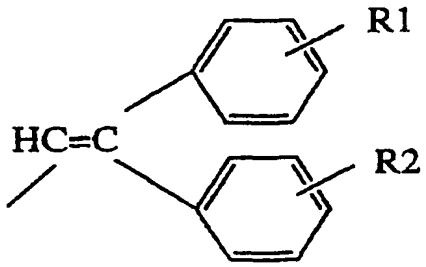


where Ar1 and Ar2 may be the same or different, and each independently represents a substituted or unsubstituted aryl group; Ar3 represents a substituted or unsubstituted arylene group; X represents a substituent containing two or more carbon rings and non-planarly bonding to a diphenylamine portion; and Y represents a substituted or unsubstituted aryl group containing five or more conjugated bonds.

14. A thin film EL device according to claim 13, wherein said compound represented by the general formula (1) has a portion contributing to luminescence where at least two molecular orbitals contributing to luminescent transition are localized.

15. A thin film EL device according to claim 13, wherein said X in the general formula (1) is a substituent represented by the following general formula (2):

(2)



where R1 and R2 may be the same or different, and each independently represents a hydrogen atom or an alkyl group.

16. A thin film EL device according to claim 13, wherein said X in the general formula (1) is a substituent represented by the following general formula (3):

where R1 and R2 may be the same or different, and each independently represents a hydrogen atom or an alkyl group.

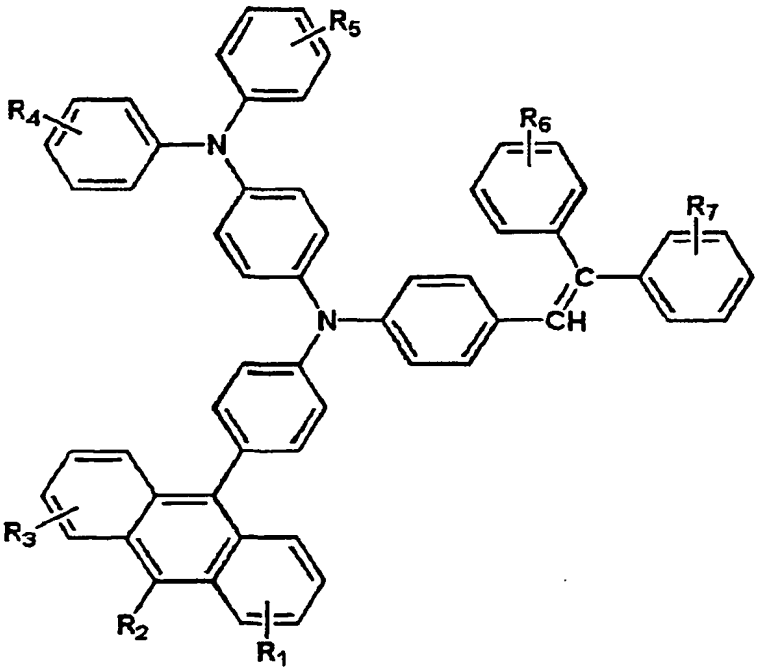
19. A thin film EL device according to claim 13, wherein said Y in the general formula (1) is an aryl group substituted with an electron-donating substituent.

20. A thin film EL device according to claim 13, wherein said Ar3 in the general formula (1) is a p-phenylene group.

21. A thin film EL device according to claim 13, wherein said Ar3 in the general formula (1) is an m-phenylene group.

22. A thin film EL device according to claim 13, wherein said hole-transport luminescent material is a compound represented by the following general formula (6):

(6)



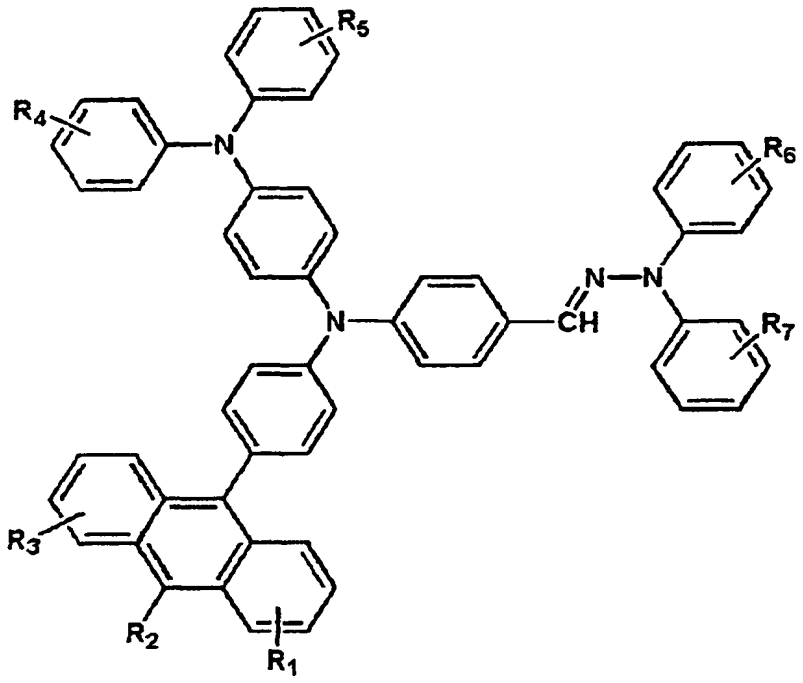
where R4, R5, R6, and R7 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group; and R1, R2, and R3 may be the same or different, and each independently represents a hydrogen atom or an electron-donating substituent.

23. A thin film EL device according to claim 22, wherein said compound represented by the general formula (6) is (4-{[4-(2,2-diphenylvinyl)phenyl] [4-(9-anthryl)phenyl]amino}phenyl)diphenylamine.

24. A thin film EL device according to claim 22, wherein said compound represented by the general formula (6) is (4-{[4-(2,2-diphenylvinyl)phenyl][4-(10-methoxy(9-anthryl))phenyl]amino}phenyl)diphenylamine.

25. A thin film EL device according to claim 13, wherein said hole-transport luminescent material is a compound represented by the following general formula (7):

(8)



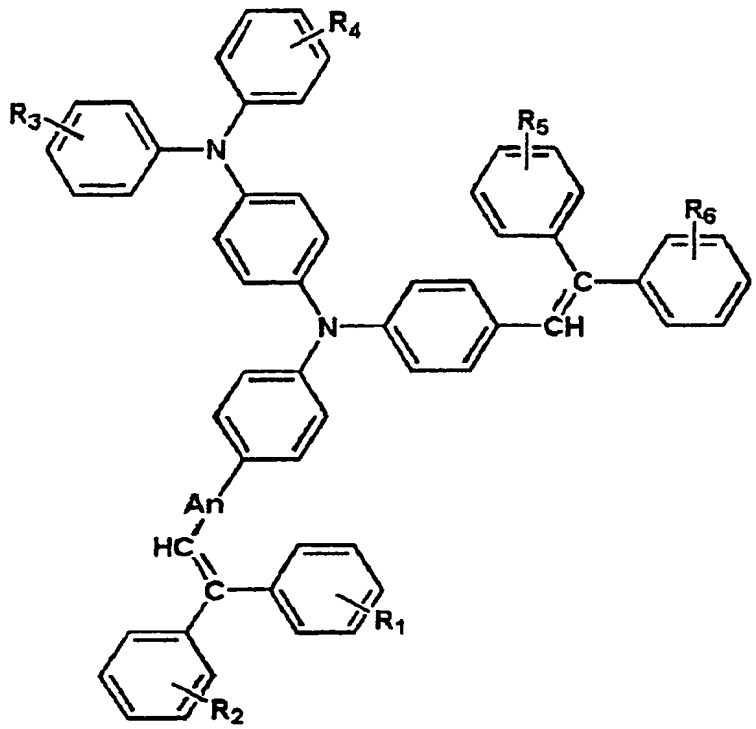
where R4, R5, R6, and R7 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group; and R1, R2, and R3 may be the same or different, and each independently represents a hydrogen atom or an electron-donating substituent.

29. A thin film EL device according to claim 28, wherein said compound represented by the general formula (8) is [4-({4-[2-aza-2-(diphenylamino) vinyl]phenyl}{4-(9-anthryl)phenyl}amino)phenyl]diphenylamine.

30. A thin film EL device according to claim 28, wherein said compound represented by the general formula (8) is [4-({4-[2-aza-2-(diphenylamino) vinyl]phenyl}{4-(10-methoxy(9-anthryl))phenyl}amino)phenyl] diphenylamine.

31. A thin film EL device according to claim 13, wherein said hole-transport luminescent material is a compound represented by the following general formula (9):

(10)



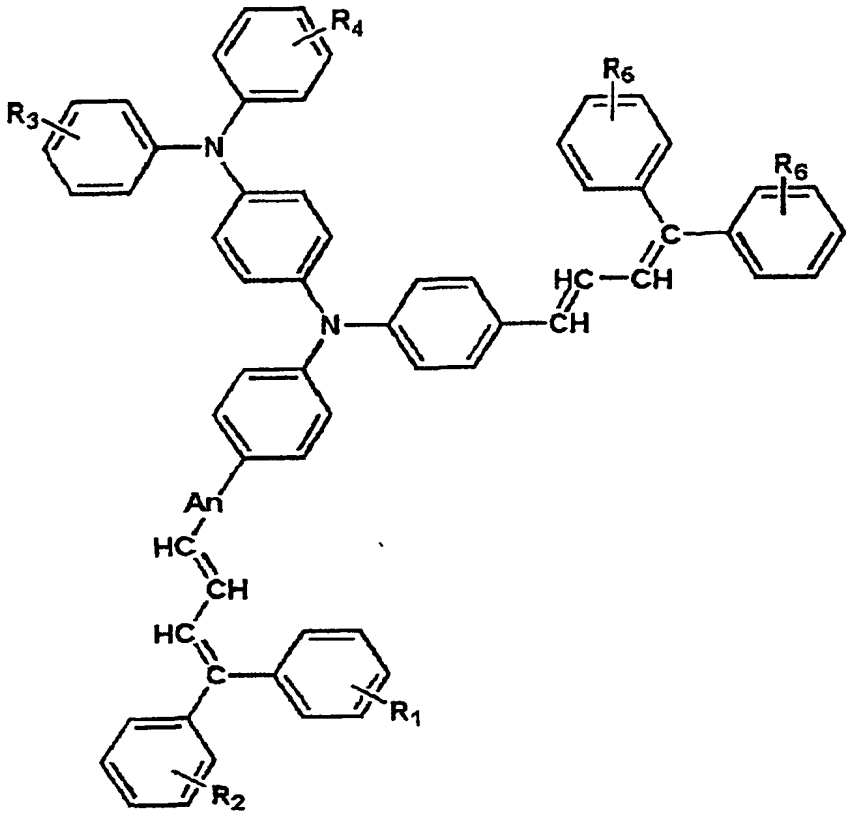
where R1, R2, R3, R4, R5, and R6 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group; and An represents an arylene group composed of two or more substituted or unsubstituted fused rings.

35. A thin film EL device according to claim 34, wherein said compound represented by the general formula (10) is [4-({4-[10-(2,2-diphenylvinyl)(9-anthryl)]phenyl}[4-(2,2-diphenylvinyl)phenyl] amino)phenyl]diphenylamine.

36. A thin film EL device according to claim 34, wherein said compound represented by the general formula (10) is [4-({4-[10-(2,2-diphenylvinyl)(9-anthryl)]phenyl}{4-(2,2-diphenylvinyl)phenyl}amino)phenyl]bis(4-methoxyphenyl) amine.

37. A thin film EL device according to claim 13, wherein said hole-transport luminescent material is a compound represented by the following general formula (11):

(12)



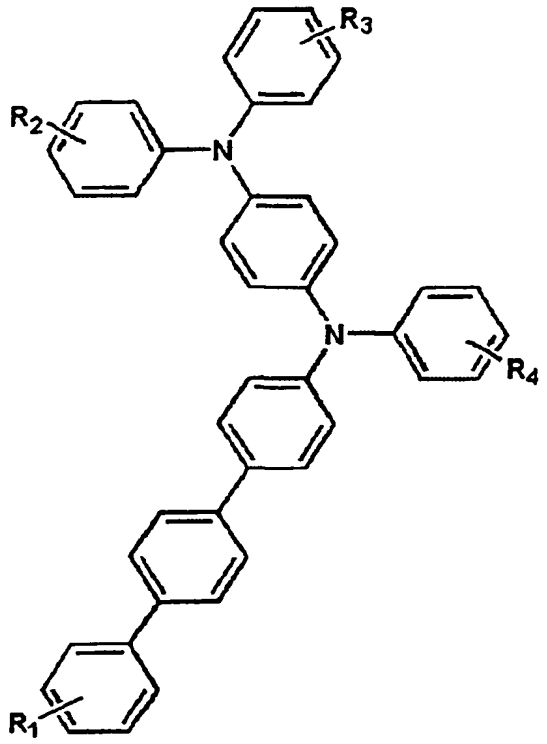
where R1 and R2 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group; and An represents an arylene group composed of two or more substituted or unsubstituted fused rings.

41. A thin film EL device according to claim 40, wherein said compound represented by the general formula (12) is [4-({4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)]phenyl}[4-(4,4-diphenylbuta-1,3-dienyl)phenyl]amino) phenyl] diphenylamine.

42. A thin film EL device according to claim 40, wherein said compound represented by the general formula (12) is [4-({4-[10-(4,4-diphenylbuta-1,3-dienyl)(9-anthryl)]phenyl}{4-(4,4-diphenylbuta-1,3-dienyl)phenyl}amino) phenyl] bis(4-methoxyphenyl)amine.

43. A thin film EL device according to claim 13, wherein said hole-transport luminescent material is a compound represented by the following general formula (13):

(14)



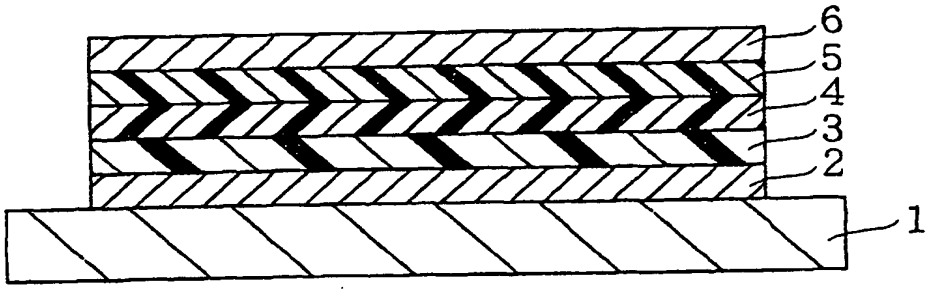
where R4 represents a hydrogen atom, an alkyl group, an alkoxy group, or an aralkyl group; and R1, R2, and R3 may be the same or different, and each independently represents a hydrogen atom, an alkyl group, or an alkoxy group.

49. A thin film EL device according to claim 48, wherein said compound represented by the general formula (14) is [4-(diphenylamino)phenyl](4-(4-phenylphenyl)phenyl]phenylamine.

50. A thin film EL device according to claim 48, wherein said compound represented by the general formula (14) is [4-{bis(4-methoxyphenyl)amino} phenyl] [4-{4-(4-methoxyphenyl)phenyl}phenyl][4-(1-methyl-1-phenylethyl) phenyl] amine.

51. A thin film EL device according to claim 13, wherein said hole-transport luminescent material is a compound represented by the following general formula (15):

Fig. 1



INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP00/09064

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
x	JP, 8-12969, A (TDK Corporation), 16 January, 1996 (16.01.96), Par. Nos. [0066], [0074] (Family: none)	1-6,12
x	JP, 8-333569, A (Idemitsu Kosan Co., Ltd.), 17 December, 1996 (17.12.96), Par. Nos. [0044], [0062] (Family: none)	1-6,12
x	JP, 11-255716, A (Sharp Corporation), 21 September, 1999 (21.09.99), Par. No. [0026] (Family: none)	1-6,12
x	JP, 10-245549, A (MINOLTA CO., LTD.), 14 September, 1998 (14.09.98), Par. No. [0018], [0026] (Family: none)	1-6,12
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